

**REMARKS**

Applicants have amended their claims in order to further clarify the definition of various aspects of the present invention. Specifically, Applicants have amended each of the independent claims in the application, that is, claims 13, 14, 20 and 24, to recite that an active material of the negative electrode is graphite, and to recite that this graphite active material of the negative electrode comprises the recited graphite powder. Note, for example, page 4, lines 19-27, of Applicants' specification.

Initially, it is respectfully requested that the present amendments be entered. Noting especially arguments previously made, for example, in the Amendment filed May 10, 2005 (see, e.g., page 10 thereof) and also noting prior recitation that the negative electrode includes the specified graphite powder, it is respectfully submitted that the present amendments do not raise any new issues, including any issue of new matter. Furthermore, noting that the present amendments further define the non-aqueous secondary battery of the present claims, including structure of the negative electrode, it is respectfully submitted that the present amendments materially limit issues remaining in connection with the above-identified application; and, at the very least, present the claims in better form for appeal. Noting additional arguments made by the Examiner in the Office Action mailed August 24, 2005, it is respectfully submitted that the present amendments are timely.

In view of the foregoing, it is respectfully submitted that Applicants have made the necessary showing under 37 CFR 1.116(c); and that, accordingly, entry of the present amendments is clearly proper.

Applicants respectfully submit that all of the claims presented for consideration by the Examiner patentably distinguish over the teachings of the prior art applied by the Examiner in rejecting claims in the Office Action mailed August 24, 2005, that is, the teachings of the U. S. Patents to Takami, et al., No. 5,340,670, and to Flandrois, et al., No. 5,554,462, under the provisions of 35 USC 102 and 35 USC 103.

It is respectfully submitted that these references as applied by the Examiner would have neither taught nor would have suggested such a non-aqueous secondary battery as in the present claims, including, inter alia, wherein an active material of the negative electrode is graphite, and this graphite active material of the negative electrode includes graphite powder having substantially completely a crystal structure, the graphite powder having a particle size equal to or smaller than 100  $\mu\text{m}$  and a deintercalating capacity for lithium of at least 320 mAh/g, as in claims 13, 14, 20 and 24; with a rhombohedral fraction, of the crystal structure of the graphite powder, being in a range of 0-20% by weight (see claim 13; note also claim 20), or wherein a hexagonal fraction, of the crystal structure of the graphite powder, is in a range of at least 80% by weight (see claim 14), or wherein a fraction of the rhombohedral crystal structure is equal to or less than 20% by weight and a fraction of the hexagonal crystal structure is equal to or more than 80% by weight (see claim 24). Note also claim 21, reciting that a fraction of hexagonal crystal structure, of the crystal structure of the graphite powder having the specified rhombohedral structure, is equal to or more than 80% by weight; and claims 32 and 33, reciting that the crystal structure of the graphite powder includes at least a fraction having hexagonal crystal structure.

The invention as claimed in the above-identified application is directed to a non-aqueous secondary battery using a negative electrode that has graphite as the active material, this graphite active material being made of graphite powder. The present invention has use, for example, in connection with a lithium battery, and provides such battery which can have a high energy density and a long life, and which has excellent intercalation capacity (that is, an increased capacity of the battery).

Carbon material has been proposed as negative electrode active material in place of lithium metal, in lithium batteries, in which charge and discharge reactions involving lithium ion intercalation into the carbon material and deintercalation from the carbon material occur. However, in previously proposed batteries (e.g., lithium secondary batteries) using carbon material as active material for the negative electrode, a large capacity cannot be obtained; and, moreover, preferable performances of rapid charging and discharging are not obtained. Note the paragraph bridging pages 2 and 3 of Applicants' specification.

Against this background, Applicants provide a non-aqueous secondary battery using a negative electrode with graphite active material, which active material has good charging and discharging characteristics, while having a large capacity (large intercalation capacity). Applicants have found that by using, as this graphite active material, graphite powder having substantially completely a crystal structure, with this crystal structure being at least 80% hexagonal crystal structure and/or at most 20% rhombohedral crystal structure, for the overall structure of the graphite powder (which graphite powder, e.g., has laminated graphite layers); and, in particular, by limiting the amount of rhombohedral crystal structure of the graphite powder and increasing the amount of hexagonal crystal structure thereof, various advantages are

achieved. In particular, by reducing the amount of rhombohedral crystal structure, of the graphite powder, and increasing hexagonal crystal structure, intercalation capacity of the negative electrode is increased. That is, the number of storage sites for lithium in the negative electrode is increased.

The present invention focuses on the crystal structure of the graphite powder of the active material of the negative electrode, requiring that the graphite powder has substantially completely a crystal structure, and the crystal structure is constituted of hexagonal crystal structure of at least 80% by weight and rhombohedral crystal structure in a range of 0-20% by weight. The graphite powder of the active material of the negative electrode, according to the present invention, has increased hexagonal crystal structure, due to an orderliness and regularity of the hexagonal-net-plane layers, and the regular stacking of these layers. That is, it is respectfully submitted that the fundamental hexagonal crystal structure, of the substantially complete crystal structure of the graphite powder according to the present invention, is achieved based upon regularity and orderliness of the lamination (stacking) of these layers. In accordance with the present invention, the existing ratio of hexagonal crystal structure, of the substantially complete crystal structure of the graphite powder as a whole, is specified at a high level, and/or the rhombohedral crystal structure is at a low level, in order to achieve the increased capacity of the battery as described in Applicants' disclosure.

In comparison, and as will be discussed in more detail infra, Takami, et al. discloses crystallites and structure with hexagonal-net-plane layers; however, this reference is silent with respect to orderliness and regularity of the laminated (stacked) hexagonal-net-plane layers. More specifically, it is respectfully submitted that Takami, et al. teaches away from such orderliness and regularity (e.g., a

substantially complete crystal structure) of the laminated hexagonal-net-plane layers, in describing that the hexagonal-net-plane layers stacked in the graphite structure “have appropriate displacements, twists and angles to one another”, so that when the stacked hexagonal-net-layers have appropriate displacements, twists, and angles to one another, lithium ions diffuse more easily between the hexagonal-net-plane layers, so the carbonaceous material presents a property of reversibly, rapidly absorbing and desorbing a large number of lithium ions. See column 7, lines 29-40.

It is respectfully submitted that the carbonaceous material in Takami, et al. is constituted by stacking “hexagonal-net-plane layers”, and each of the layers is constituted by a unit plane layer A, B or C and each unit plane layer is constituted by a large number of crystallites spread in a plane, each of which is constituted by 6 carbon atoms, as, for example, illustrated in Figs. 1 and 3 of Flandrois, et al.

Importantly, it is respectfully submitted that Takami, et al. nowhere discloses the stacking condition of the hexagonal-net-plane layers, other than describing that the layers stacked in the graphite structure “have appropriate displacements, twists and angles to one another”. As stated previously, and discussed further infra, such appropriate displacements, twists and angles to one another teach away from the substantially complete crystal structure of the graphite powder as in the present claims. Specifically, according to the present invention, the layers are stacked in an orderly and regular manner to form substantially complete crystal structure in the graphite.

While Takami, et al. describes crystal planes (faces) (101) and (100) for specifying the intensity ratio  $P_{101}/P_{100}$  of the (101) diffraction peak  $P_{101}$  to the (100) diffraction peak  $P_{100}$  of the graphite structure, Takami, et al. refers to crystallites of the graphite structure, and it is respectfully submitted that this reference does not

identify crystal planes, or whether the planes relate to hexagonal crystal structure or rhombohedral crystal structure.

It is respectfully submitted that Takami, et al. specifically discloses the need for twisted structures, thus having a number of twisted structures. In comparison, the present invention has a substantially complete crystal structure, substantially without twisted structures as required by Takami, et al. As contended previously, and as will be discussed further infra, it is respectfully submitted that the disclosure of Takami, et al. in connection with “displacements, twists and angles” of the hexagonal-net-plane layers to one another would have taught away from the substantially complete crystal structure with the recited hexagonal/rhombohedral crystal structure, and advantages thereof with respect to increased intercalation capacity of the negative electrode, as in the present invention.

Furthermore, and as indicated previously, Takami, et al. discloses crystallites, and structure with hexagonal-net-plane layers which are laminated; however, it is respectfully submitted that this reference is silent with respect to orderliness and regularity of the laminated (stacked) hexagonal-net-plane layers; and, in particular, it is respectfully submitted that this reference is silent in connection with the substantially complete crystal structure. In connection with differences between crystallites and crystals, attention is again respectfully directed to the definition of “crystal” and of “crystallite” on page 327 of Hawley’s Condensed Chemical Dictionary (12<sup>th</sup> Ed. 1993), submitted with the Submission (Amendment) filed November 22, 2004. As can be appreciated therefrom, a crystallite is that portion of a crystal whose constituent atoms, ions or molecules form a perfect lattice, without strains or other imperfections, and is usually microscopic (while crystals may be quite large). Taking the disclosure of Takami, et al. as a whole, including the disclosure therein of

“appropriate displacements, twists and angles” of the planar layers to one another (note, for example, column 7, line 21-40 of Takami, et al.), it is respectfully submitted that Takami, et al. would have neither disclosed nor would have suggested, and in fact would have taught away from, the crystal powder having a substantially complete crystal structure, and especially with specified fractions of the crystal structure being hexagonal and/or rhombohedral, as in the present claims, and advantages thereof.

In the first full paragraph on page 3 of the Office Action mailed August 24, 2005, the Examiner contends that a material would necessarily have to comprise an entire crystal structure “to exhibit such analysis when undergoing X-ray defraction procedures”. The Examiner provided no evidence or reasoning in support of this contention. Absent evidence or reasoning in support thereof, it is respectfully submitted that this contention by the Examiner is clearly improper under the requirements of 35 USC 102 and 35 USC 103. See In re McKellin, 188 USPQ 428 (CCPA 1976). In this regard, it is again emphasized that Takami, et al. refers to hexagonal-net-plane layers.

The acknowledgement that the Examiner attributes to Applicants, in the last 3 lines of the first paragraph on page 3 of the Office Action mailed August 24, 2005, is noted. It is respectfully submitted that Applicants acknowledge that Takami, et al. describes an intensity ratio of a graphite structure, e.g., in column 3, lines 1-8; and also describes a mean length La of a crystallite in an a-axis direction of the graphite structure, e.g., in column 3, lines 15-24. Applicants do not acknowledge, and in fact respectfully traverse, any contention by the Examiner that such spectra in Takami, et al. shows a substantially complete crystal structure.

To emphasize, in accordance with the present invention, graphite powder of a relatively small size is used as the active material for the negative electrode, and a focus thereof is on a substantially complete crystallinity of this graphite powder and the type of crystal structure. Applicants have found that by using graphite powder having a substantially complete crystal structure, and having a relatively large amount of this crystal structure being hexagonal crystal structure; and, in particular, with this crystal structure of the graphite powder having a specific ratio of hexagonal crystal structure to rhombohedral crystal structure, improvements in capacity are achieved. That is, according to the present invention the graphite powder which is the active material of the negative electrode has a substantially complete graphite crystal structure, and Applicants limit the amount of rhombohedral structure and increase amount of hexagonal crystal structure, with orderliness and regularity between the laminate layers of the graphite (e.g., a substantially complete crystal structure), so as achieve unexpectedly better results of increased capacity, as seen in Applicants' disclosure.

In the first full paragraph on page 3 of the Office Action mailed August 24, 2005, the Examiner points to the recited exothermic peak and intensity ratio of two different diffraction peaks, obtained by X-ray diffraction analysis, in Takami, et al., and contends that these values indicate a crystal structure for the entire anode material, the Examiner contending that a material would necessarily have to comprise an entire crystal structure to exhibit such analysis when undergoing X-ray diffraction procedures. The Examiner has pointed to no evidence or reasoning supporting this contention. Moreover, it is respectfully submitted that this contention by the Examiner is totally inconsistent with express teachings of Takami, et al., of graphite structure having "appropriate displacements, twists and angles to one



another". From the express teachings of Takami, et al., of crystallites, wherein the stacked hexagonal-net-plane layers have appropriate displacements, twists and angles to one another, such rebuts the conclusion by the Examiner as to a crystal structure for the entire anode material in Takami, et al.

In the third paragraph on page 4 of the Office Action mailed August 24, 2005, the Examiner acknowledges that Takami, et al. has displacements, twists and angles "to some degree", but contends that Applicants have not included claim language distinguishing therefrom. This contention by the Examiner is respectfully traversed. Rather than having displacements, twists and angles "to some degree", as alleged by the Examiner, it is respectfully submitted that these displacements, twists and angles are necessary requirements of Takami, et al., to achieve diffusion of lithium ions with ease. Moreover, it is respectfully submitted that Applicants provide language distinguishing from Takami, et al., for example, in reciting that the graphite powder has substantially completely a crystal structure, as in the present claims and as described in Applicants' specification.

Takami, et al discloses a negative electrode carbonaceous material suitable for a lithium secondary battery, as well as a battery using this negative electrode carbonaceous material. According to Takami, et al., the negative electrode contains a carbonaceous material which has an exothermic peak at 700° C or more when measured by a differential thermal analysis, and an intensity ratio  $P_{101}/P_{100}$  of a (101) diffraction peak  $P_{101}$  to a (100) diffraction peak  $P_{100}$  of a graphite structure obtained by X-ray diffraction analysis, of 0.7-2.2; and absorbs and desorbs lithium ions. See column 3, lines 1-8. Other negative electrodes disclosed in Takami, et al., are described at column 3, lines 16-25, 38-44, 56-60 and 66-68; as well in column 4, lines 9-13, 17 and 18.

Note also column 20, lines 25-56; column 21, lines 3-22; column 22, lines 1-19 and 37-62; and column 23, lines 10-21, for other disclosures in connection with negative electrodes of the secondary battery of Takami, et al. These disclosures all refer to the carbonaceous material having a graphite structure that exhibits a property of allowing reversible absorption and desorption of lithium ions to and from hexagonal-net-plane layers in the graphite structure. Note also, for example, example 1 in column 24 and particularly lines 23-29 thereof, disclosing that the carbonaceous material used was a graphitized carbon powder with an average particle size of 25  $\mu\text{m}$ , which was distributed at a ratio of 90 vol.% within the range of 1-50  $\mu\text{m}$  in a particle size distribution, and in which the ratio of particles with a particle size of 0.5  $\mu\text{m}$  or less was 0 vol. % in the particle size distribution. Note also, inter alia, example 2 and the discussion in connection therewith at column 25, lines 20-26; and example 3 and the discussion in connection therewith at column 25, lines 52-59, describing particle size of the carbonaceous material.

Attention is particularly directed to column 7, lines 29-40, of Takami, et al., emphasizing that the carbonaceous material having the recited intensity ratio has appropriate displacements, twists and angles of the hexagonal-net-plane layers so that lithium ions diffuse more easily between the hexagonal-net-plane layers; and note also column 14, lines 21-39, disclosing that the carbonaceous material has a mean length La of a crystallite in the a-axis direction of graphite structure. This patent again refers to crystallite structure, e.g., at column 22, lines 37-62.

It is respectfully submitted that the disclosure in Takami, et al., having, e.g., crystallites and having hexagonal-net-plane layers with appropriate displacements, twists and angles, would have neither disclosed nor would have suggested the presently claimed structure, including the graphite powder having substantially

completely the crystal structure, with the recited particle size of the graphite powder and with specified hexagonal and/or rhombohedral fraction of the crystal structure of the graphite powder, and the graphite powder having a deintercalating capacity for lithium of at least 320 mAh/g, and advantages thereof, as described in the foregoing.

The contention by the Examiner on page 2 of the Office Action mailed August 24, 2005, that it is inherent in the teachings of Takami, et al. that the capacity for the graphite crystal powder would be at least 320 mAh/g, is respectfully traversed. Initially, contrary to the conclusion by the Examiner, it is respectfully submitted that Takami, et al. does not teach the same material as in the present claims. It is respectfully submitted that the graphite in Takami, et al. would not have the crystal structure of the present invention. In this regard, note that Takami, et al. uses, as starting materials, carbonaceous material such as mesophase pitch, mesophase spherulites and coke. In contrast, the present invention uses natural graphites.

In addition, it is again emphasized that Takami, et al. discloses materials having hexagonal-net-plane layers stacked so as to have displacements, twists and angles to one another. Clearly, the material in Takami, et al. is not the same as that of the present invention, and it is respectfully submitted that such material in Takami, et al. would not inherently have the deintercalating capacity recited in the present claims.

It is again noted that, with regard to planes (faces) (100) and (101) in the intensity ratio of  $P_{101}/P_{100}$ , Takami, et al. gives no definition with respect to crystal planes in the hexagonal crystal structure and in the rhombohedral crystal structure. Assuming, arguendo, that plane (100) in Takami, et al. is of hexagonal structure and plane (101) is of rhombohedral structure, described in the above-identified

application, it is respectfully submitted that the intensity ratio of  $P_{101}/P_{100}$  in a range of 0.7-2.2 in Takami, et al. indicates an inclusion of hexagonal crystal structure contained in the graphite structure being less than 80% by weight.

For all of the foregoing reasons, it is respectfully submitted that Takami, et al. would have neither taught nor would have suggested, and in particular would have taught away from, the presently claimed non-aqueous secondary battery, wherein the negative electrode thereof includes graphite as active material thereof, and the graphite active material of the negative electrode is graphite powder having substantially completely a crystal structure, with hexagonal and/or rhombohedral fractions thereof and particle size thereof, the graphite powder having the recited deintercalating capacity, and advantages thereof.

Flandrois, et al. discloses a carbon anode for a lithium rechargeable electrochemical cell, and a process for its production. The carbon anode comprises a graphite-containing, carbon-containing material, characterized in that the material includes, prior to electrical cycling, at least a first phase constituted by graphite having a rhombohedral crystal structure and comprising a fraction of more than 10%. See column 2, lines 3-10. Note also column 2, lines 33-40. See also the examples and Tables in connection therewith, showing, inter alia, passivation capacity (mAh/g).

Initially, it is noted that Flandrois, et al. discloses a carbon anode which includes a graphite-containing, carbon-containing material. Such disclosure of an anode would have neither disclosed nor would have suggested the presently claimed subject matter, including the negative electrode having an active material thereof being graphite, the graphite active material of the negative electrode including the specified graphite powder. Clearly, the carbon anode is the positive electrode in

Flandrois, et al. Moreover, it is respectfully submitted that Flandrois, et al. is concerned with providing a carbon-containing anode having increased rhombohedral structure, so as to increase passivation capacity. In contrast, the graphite powder of the negative electrode of the present invention has increased hexagonal crystal structure and decreased rhombohedral crystal structure so as to increase deintercalating capacity for lithium. It is respectfully submitted that Flandrois, et al. would have neither taught nor would have suggested the presently claimed invention, including, inter alia, ranges of hexagonal and rhombohedral fractions, and unexpectedly better results achieved in these ranges, as in the present invention; as well as other features of the present invention as discussed previously.

In particular, it is respectfully submitted that Flandrois, et al. would have neither taught nor would have suggested the deintercalating capacity for lithium as in the present claims. In connection therewith, it is noted that Flandrois, et al. refers to a passivation capacity  $\Delta$ , in the Tables associated with the various examples and comparative examples. As can be seen, for example, in Fig. 7 of Flandrois, et al., it is respectfully submitted that " $\Delta$ " is the distance between the curves 71 and 72. In contrast, it is respectfully submitted that the deintercalating capacity according to the present invention is between curve 72 in Fig. 7 of Flandrois, et al. and the point where curves 71 and 72 intersect the abscissa, as can be seen in the copy of Fig. 7 of Flandrois, et al. enclosed with the Amendment filed May 10, 2005. It is respectfully submitted that the passivation capacity as in Flandrois, et al. would have neither taught nor would have suggested the deintercalating capacity for lithium, of the graphite powder, according to the present invention, and advantages thereof.

The contention by the Examiner in the last 3 lines of the second paragraph of Item 5, on page 5 of the Office Action mailed August 24, 2005, is noted. Contrary to

the conclusion by the Examiner, it is respectfully submitted that Flandrois, et al. does not teach "the same material" as in Applicants' claims, and that the graphite powder in Flandrois, et al. would not inherently have the recited deintercalating capacity for lithium as recited in the present claims. Moreover, noting that Flandrois, et al. discloses the material as forming the anode, clearly Flandrois, et al. would have neither taught nor would have suggested the presently claimed structure, including the negative electrode.

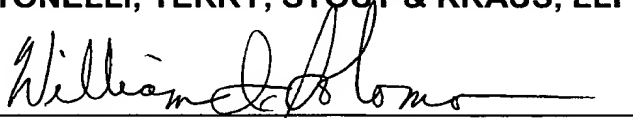
In view of the foregoing comments and amendments, entry of the present amendments, and reconsideration and allowance of all claims presently pending in the above-identified application, are respectfully requested.

Please charge any shortage in the fees due in connection with the filing of this paper to the Deposit Account of Antonelli, Terry, Stout & Kraus, LLP, Deposit Account No. 01-2135 (case No. 503.34465VV4), and please credit any excess fees to such Deposit Account.

Respectfully submitted,

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